

**Final Technical Progress Report  
to the  
Air Force Office of Scientific Research  
for research on  
NON-LINEAR MICROWAVE AND  
FAST OPTICS STUDIES OF  
YBCO SUPERCONDUCTING FILMS**

**AFOSR Agreement #F49620-96-1-0392  
for the period  
August 1, 1996 — February 28, 1999**

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August 16, 2000**

**20000908 059**

**DTIC QUALITY INSPECTED 4**

## REPORT DOCUMENTATION PAGE

AFRL-SR-BL-TR-00-

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for review the data needed, and completing and reviewing this collection of information. Send comments regarding this burden estimate or reducing this burden to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503

 Submitting  
 for  
 use of

1. AGENCY USE ONLY (Leave blank)

2. REPORT DATE  
August 16, 2000

3. REPORT TYPE AND DATES COVERED

August 1, 1996-February 28, 1999

FINAL

4. TITLE AND SUBTITLE

Non-Linear Microwave and Fast Optics Studies of YBCO Superconducting Films

5. FUNDING NUMBERS

F49-620-96-1-0392

6. AUTHOR(S)

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7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

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REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)

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Arlington, VA 22203-197710. SPONSORING / MONITORING  
AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION / AVAILABILITY STATEMENT

**DISTRIBUTION STATEMENT A**  
 Approved for Public Release  
 Distribution Unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 Words)

During the period of this contract, the research program, "Non-linear microwave and fast optics studies of YBCO superconducting films" focused on several projects central to the program objectives. The main focus was on a collaborative study of the nonlinear microwave properties of superconducting YBCO, for which the research was carried out with collaborators at the Air Force Research Laboratory at Hanscom Air Force Base and the MIT Lincoln Laboratory. The research work included studies of: (1) the effect of the angular dependence of grain boundaries on the linear and non-linear microwave impedance of YBCO, (2) an extension of the coupled grain model to account for the distributed properties of junctions that are much longer than the London penetration depth, and have a distribution of the critical current densities and the normal state resistivities along the junction length, (3) second harmonic generation by Josephson junctions, (4) the effect of patterning on the surface impedance of YBCO thin films, and (5) the use of fast optics to investigate the basic interaction responsible for the high  $T_c$  superconducting state in the BCS alkali metal-doped fullerene superconductors of  $K_3C_{60}$  and  $Rb_3C_{60}$ . During this period one graduate student completed his M.S. thesis and three students completed Ph.D. theses.

14. SUBJECT TERMS

Superconductors, microwave properties, Josephson junctions, fast optics, vortex dynamics,

15. NUMBER OF PAGES

10

16. PRICE CODE

17. SECURITY CLASSIFICATION  
OF REPORT

UNCLASSIFIED

18. SECURITY CLASSIFICATION  
OF THIS PAGE

UNCLASSIFIED

19. SECURITY CLASSIFICATION  
OF ABSTRACT

UNCLASSIFIED

20. LIMITATION OF ABSTRACT

NSN 7540-01-280-5500

 Standard Form 298 (Rev. 2-89)  
 Prescribed by ANSI Std. Z39-18  
 298-102

DTIC QUALITY INSPECTED 4

## OBJECTIVES

### Statement of Work

1. Correlation of the dc transport properties of thin YBCO films with the non-linear power dependence of the surface impedance.
2. Characterization of the YBCO films used for microwave studies by a variety of techniques such as scanning tunneling microscopy, atomic force microscopy, and x-ray diffraction.
3. Study of the non-linear rf power dependence of the surface impedance of YBCO thin films in the low frequency limit.
4. Investigation of the non-linear properties of thin-film Josephson junctions.
5. Identification of loss mechanisms in high- $T_c$  films in order to increase power handling capability of the materials and of microwave devices made from these materials.
6. Investigation of the fast-optical properties of thin YBCO films with a Ti:sapphire laser utilizing recent advances in pump-probe techniques.

### STATUS OF EFFORT

During the third period of this contract (1998-99), the research program on "Non-linear microwave and fast optics studies of YBCO superconducting films" focused on several projects central to the program objectives. The main focus was a collaborative study of the nonlinear microwave properties of superconducting YBCO, for which the research was carried out with collaborators at the Air Force Research Laboratory at Hanscom Air Force Base and the MIT Lincoln Laboratory. The research work included studies of: (1) the effect of the angular dependence of grain boundaries on the linear and non-linear microwave impedance of YBCO, (2) an extension of the coupled grain model to account for the distributed properties of junctions that are much longer than the London penetration depth, and have a distribution of the critical current density and the normal state resistivity, (3) second harmonic generation by Josephson junctions, (4) the effect of patterning on the surface impedance of YBCO thin films, and (5) the use of fast optics to investigate the basic interaction responsible for the high  $T_c$  superconducting state in the BCS alkali metal-doped fullerene superconductors of  $K_3C_{60}$  and  $Rb_3C_{60}$ . During this period one graduate student completed his M.S. thesis.

## ACCOMPLISHMENTS/NEW FINDINGS

### Microwave Properties of High $T_c$ Superconductors

During the 1997-98 year, our group worked on several research projects of relevance to the microwave properties of high  $T_c$  superconducting thin films. The accomplishments and new findings for each project are summarized below. The research projects described here were carried out in close collaboration with researchers at the Air Force Research Laboratory at Hanscom Air Force Base and Lincoln Laboratory. Extensive use was made of the facilities at both locations, as well as facilities on the MIT campus. The objective of this research is to gain a better understanding of the microwave-frequency power dependence of the surface impedance in high-temperature superconducting YBCO thin films in order to improve their power handling capabilities.

### Angular Dependence of Grain Boundaries in Engineered Josephson Junctions

Microwave frequency Josephson junction effects were studied using Josephson junctions produced by grain boundaries that were introduced by depositing YBCO thin films on sapphire bicrystal substrates with a variety of misalignment angles at the bicrystal interface. This project had been initiated during the previous year. The first experiments were detailed studies for a sample prepared with a misalignment angle of  $24^\circ$ , to identify the various types of phenomena that are observed. These detailed measurements showed nonlinearities in the microwave impedance especially a step behavior both in the resistance and the reactance due to the grain boundary. The steps observed in the resistance as a function of rf current were analyzed in detail. A long junction circuit model was developed, and was used to calculate the resistance and reactance as a function of rf current. The model results were found to agree very well with the measurements. The modeling specifically showed that the measured steps in the resistance were due to Josephson vortices created by rf currents, with each step corresponding to an additional Josephson vortex being generated per rf cycle as the current is increased. This work was published in Physical Review B.

The new studies that were initiated this year extend the measurement of the microwave impedance of engineered grain boundaries to YBCO films deposited on sapphire bicrystal substrates with different misorientation angles  $\theta$  ranging from 0 to 24 degrees, including  $\theta = 2^\circ, 5^\circ, 10^\circ$  and  $24^\circ$  samples. Typically, YBCO films have misorientation angles between grains of only 2 to 5 degrees. The data are compared with measurements made on YBCO samples prepared on single-crystal substrates. The results of rf measurements show that engineered grain boundaries of 5 degrees or less

have essentially no effect on microwave power handling, and misorientation angles as high as 10 degrees have a relatively small effect. The dc measurements were performed on a four-point test structure on the same substrate as the resonator. Although the measurements demonstrate that low-angle grain boundaries ( $\theta \leq 10^\circ$ ) have little effect on the rf power handling, grain boundaries with angles as low as  $5^\circ$  are sufficient to cause significant nonlinear dc losses due to Josephson vortices. These small angle grain boundaries have almost no effect on the rf critical currents. Further, even when the grain boundaries do reduce the rf critical current, the relative effect is always more than an order of magnitude greater for the dc case than for the case at microwave frequencies. The results of this study have important implications for the production of YBCO thin films with greater rf power handling capabilities than those presently manufactured. This work has been accepted for publication in Applied Physics Letters.

### **Non-linear Long-junction model for the microwave impedance**

A circuit model has been developed for Josephson junctions (JJ) that solves the nonlinear long-junction equation, driven by a nonuniform current distribution. This extended resistively shunted junction (ERSJ) model consists of a parallel array of ideal resistively shunted JJs connected by inductors. The junction array is connected to an array of current sources that simulate the time- and space-dependent current distribution in a stripline. This model describes the creation, annihilation and motion of Josephson vortices and explains the experimentally measured step structure in the power dependence of the effective resistance in Y-Ba-Cu-O Josephson junctions. The calculated reactance also fits the experimental data. This model thus contributes to a better understanding of the power-handling characteristics of high- $T_c$  microwave devices, in which the power losses are believed to result from Josephson-junction effects associated with imperfections in the films. The model also predicts second-harmonic generation with a highly nonlinear and non-monotonic power dependence. Details of the dynamics of Josephson vortices are presented and discussed in a paper submitted to the Journal of Superconductivity.

### **Second Harmonic Generation**

The generation of second harmonics by Josephson junctions was predicted theoretically by the modeling results of Chris Lehner on the long junctions. To this end, Joe Habib set up an experiment to measure the generation of second harmonics in YBCO thin films with and without grain boundaries. Harmonics generation may shed some new light on the underlying mechanism causing the non-linearity in the power handling of YBCO thin films at microwave frequencies. Large amplitude harmonics have been ob-

served. However, the measurements are still at an early stage and the data interpretation is incomplete at this time.

### **Effect of Patterning on the Surface Impedance of YBCO Thin Films**

We have made measurements of the surface impedance of YBCO thin films using various techniques, including the stripline resonator and dielectric resonator techniques. The main purpose of the experiments is to investigate the difference in power dependence of patterned and unpatterned YBCO thin films, since there have been suggestions that most of the nonlinear effects in the surface impedance of YBCO thin films are due to patterned edge effects. The surface resistance at microwave frequency (rf) of unpatterned YBCO thin films deposited on a 2 inch-diameter  $\text{LaAlO}_3$  wafers was measured using a sapphire dielectric-resonator technique at 10.7 GHz. The surface resistance of Nb and gold films was also measured to validate this technique. The rf measurements were made as a function of peak rf magnetic field (0.1 to 100 Gauss) and temperature (30 to 80K). In addition to the standard frequency-domain method to measure the surface resistance of YBCO films, a time-domain method with pulsed-rf input was employed to reduce the heating effects at high input power (up to 40 dBm). The same characterized YBCO wafers were then patterned to make stripline resonators. The microwave surface impedance of the patterned films was measured using the stripline resonator technique for a wider range of peak rf magnetic field (up to 1000 Gauss), over the same temperature range as the unpatterned YBCO thin films. Preliminary experimental results show agreement between the power dependence of the surface resistance of the unpatterned and patterned YBCO thin films, suggesting that the patterning had little effect on the surface resistance under the measurement conditions.

### **Distributed Resistively Shunted Junction Model**

Work has also been done on the coupled-grain/resistively shunted junction (RSJ) modeling of YBCO thin films (Distributed RSJ Model). We have modeled YBCO thin films in terms of a series of ideal superconducting grains, coupled by resistively shunted Josephson junctions which simulate the grain boundaries. Independent distributions for the critical current  $I_c$  and the normal state resistance  $R_n$  of the junctions have been incorporated in order to simulate YBCO thin films with a distribution of defects, corresponding, for example, to grain boundaries with a range of grain boundary angles. Due to the intensive computational requirements of this model, we have utilized a DoD MSRC supercomputing system. We calculate the surface resistance and reactance of YBCO thin films. Comparisons between the model and experimental results from YBCO thin-film stripline resonators

show good qualitative agreement for the resistance, for frequencies in the range 1–20 GHz. We have also modeled a phase-shifting device consisting of 300 high- $T_c$  Josephson junctions in series along a microstrip transmission line. Computed results are in good agreement with the measured device response versus microwave input power. Some work has been started on extensions of the model to include two-dimensional arrays of junctions which would be a combination of the present distributed-RSJ model and the extended-RSJ model developed previously by Chris Lehner in our group.

We have attempted to investigate the effects of introducing external pinning sites on the microwave impedance of YBCO thin films.

### Coherent phonons in alkali metal-doped $C_{60}$

An experimental study has been made of the femtosecond pump-probe reflectivity spectra in  $K_3C_{60}$  and  $Rb_3C_{60}$  as the sample temperature is lowered through the superconducting transition temperature. An analysis of the data is underway, based on the BCS theory of superconductivity. For  $K_3C_{60}$ , a complex dielectric function given by Iwasa is used to obtain an expression for  $\Delta R(0)/R$ , the initial fractional change in reflectivity immediately after the arrival of the pump pulse. The result obtained experimentally is consistent with the frequency dependence of the sign of  $\Delta R(0)/R$  observed at room temperature in  $K_3C_{60}$ . The temperature dependence of  $\Delta R(0)/R$  for  $K_3C_{60}$  calculated with the model above and below  $T_c$  is qualitatively consistent with the experimental results obtained with a probe frequency of 0.64 eV, but only if the BCS theory is used, and only if interband transitions originating at points in the conduction band below the Fermi level are included in the calculation. The experimental results for  $\Delta R(0)/R$  in  $Rb_3C_{60}$ , which are dissimilar from those for  $K_3C_{60}$ , are also consistent with a modified model for  $\Delta R(0)/R$  in which the BCS theory is taken into account, but in this case the model includes an interband component associated with states in the conduction band lying above the Fermi level.

## PERSONNEL SUPPORTED

The program involves the personnel listed in this section.

- Mildred Dresselhaus - Principal Investigator (10%). Responsible for the research and the direction of all aspects of the program.
- Gene Dresselhaus - Co-principal Investigator (25%). Responsible together with the principal investigator for the research and the direction of all aspects of the program.
- Daniel Oates - Visiting Research Scientist (supported 50% by another contract F49620-98-1-0021 under the same AFOSR program of Dr.

Harold Weinstock). Responsible together with the principal investigators for the microwave research on the high  $T_c$  thin films.

- Nathan Belk - Research Assistant, Department of Physics. Responsible for microwave studies on striplines as a function of temperature, magnetic field and frequency. He completed his Ph.D. Thesis in September 1996.
- Siegfried Fleischer - Research Assistant, Department of Electrical Engineering and Computer Science. Responsible for studies on fullerenes using pump-probe femtosecond reflectivity technique. He completed his Ph.D. Thesis in February 1997.
- Youssef Habib - Research Assistant, Department of Physics. Responsible for microwave studies on striplines containing engineered Josephson junctions.
- Boris Pevzner - Research Assistant, Department of Electrical Engineering and Computer Science (supported by another grant). Responsible for sample preparation of films of  $C_{60}$  and alkali metal doped  $C_{60}$ . These films were used in Siegfried Fleischer's thesis research.
- Hao Xin - Graduate Student, Department of Physics. Responsible for continuation of the studies of flux dynamics in YBCO films and for extending the coupled grain model.
- Chris Lehner - Graduate Student, Department of Physics and Captain, US Army on assignment at MIT. Responsible for the development of computer modeling for the Josephson junction stripline studies and experimental measurements pertinent to his models.

## PUBLICATIONS

### Theses Completed

1. Nathan Belk, "Electronic Transport and Magnetic Properties of Disordered High  $T_c$  Materials", Ph.D. Thesis, Department of Physics, Massachusetts Institute of Technology, September, 1996.
2. Siegfried Fleischer, "Ultrafast dynamics studies on fullerene thin films", Ph.D. Thesis, Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, February, 1997.
3. Christopher J. Lehner, "Modeling and Measurement of Microwave Effects in High- $T_c$  Long Josephson Junctions", M.S. Thesis, Department of Physics, Massachusetts Institute of Technology, February, 1998.



4. Youssef M. Habib, "Measurements and modeling of the microwave impedance in high- $T_c$  grain boundary Josephson junctions: Josephson fluxon generation and vortex dynamics"

## Microwave Superconductivity Studies

1. Y. M. Habib, D. E. Oates, G. Dresselhaus, and M. S. Dresselhaus, Power dependence of microwave  $Z_s$  in high- $T_c$  Josephson junctions: measurements and modeling, IEEE Trans. Appl. Superconductivity 7 2553 (1996). Proceedings of the Applied Superconductivity Conference.
2. Nathan Belk, D. E. Oates, D. A. Feld, G. Dresselhaus, and M. S. Dresselhaus, Linear and nonlinear microwave dynamics of vortices in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films, Phys. Rev. B 56, 11966-11978 (1997).
3. Y. M. Habib, C. J. Lehner, D. E. Oates, L. R. Vale, R. H. Ono, G. Dresselhaus, and M. S. Dresselhaus, Measurements and modeling of the microwave impedance in high- $T_c$  grain boundary Josephson junctions: Josephson fluxon generation and vortex dynamics, Phys. Rev. B 57, 13833-13844 (1998).
4. Y. M. Habib, D. E. Oates, L. R. Vale, R. H. Ono, G. Dresselhaus, and M. S. Dresselhaus, Microwave power handling in engineered  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  grain boundaries, Appl. Phys. Lett. (1998), accepted for publication.
5. C. J. Lehner, D. E. Oates, Y. M. Habib, G. Dresselhaus, and M. S. Dresselhaus, Modeling of the microwave impedance in high- $T_c$  long Josephson junctions, J. Superconductivity (1998), submitted for publication.

## Fullerene Studies

1. S. B. Fleischer, B. Pevzner, D. J. Dougherty, E. P. Ippen, M. S. Dresselhaus, and A. F. Hebard, Phototransformation in visible and near-IR femtosecond pump-probe studies of  $\text{C}_{60}$  films, Appl. Phys. Lett. 69, 296-298 (1996).
2. S. B. Fleischer, B. Pevzner, D. J. Dougherty, H. J. Zeiger, G. Dresselhaus, M. S. Dresselhaus, E. P. Ippen, and A. F. Hebard, Coherent phonons in alkali metal-doped  $\text{C}_{60}$ , Appl. Phys. Lett. 71, 2734-2736 (1997).
3. S. B. Fleischer, B. Pevzner, D. J. Dougherty, E. P. Ippen, M. S. Dresselhaus, and A. F. Hebard, Phototransformation in visible and near-IR

femtosecond pump-probe studies of  $C_{60}$  films, Appl. Phys. Lett. **71**, 1277 (1997).

4. S. B. Fleischer, B. Pevzner, D. J. Dougherty, H. J. Zeiger, G. Dresselhaus, M. S. Dresselhaus, E. P. Ippen, and A. F. Hebard, Fast optics in superconducting alkali-doped  $C_{60}$ , Phys. Rev. B **62**, 1366-1378 (2000).

### Conference Presentations

1. Coupled-Grain/RSJ Series Array for Modeling of Nonlinear Microwave Surface Impedance of YBCO Thin Films (Applied Superconductivity Conference, ASC 98).
2. Comparison of the microwave frequency power dependence of the surface resistance of unpatterned and patterned YBCO thin films (Materials Research Society, MRS Fall 98).
3. Measurements and Modeling of the Microwave Impedance in High- $T_c$  Grain Boundary Josephson Junctions: Josephson Fluxon Generation and Vortex Dynamics. Y. M. Habib, C. J. Lehner, D. E. Oates, L. R. Vale, R. H. Ono, G. Dresselhaus, and M. S. Dresselhaus, Oral Presentation, Materials Research Society, Boston MA, December 1998.
4. Properties of Grain-Boundary Josephson Junctions at Microwave Frequencies. Y. M. Habib, C. J. Lehner, D. E. Oates, L. R. Vale, R. H. Ono, G. Dresselhaus, and M. S. Dresselhaus, Oral Presentation, American Physical Society, Kansas City MO, March 1998.
5. Measurements and Modeling of Microwave Impedance of High- $T_c$  Grain Boundaries. D. E. Oates, Y. M. Habib, C. J. Lehner, L. R. Vale, R. H. Ono, G. Dresselhaus, and M. S. Dresselhaus, Invited Talk, Applied Superconductivity Conference, Palm Desert CA, September 1998.

## INTERACTIONS AND TRANSITIONS

### Consultative and Advisory Functions

1. Trustee, Caltech and member of the Jet Propulsion Laboratory Advisory Committee.
2. Consultant, Lawrence Livermore National Laboratory
3. Member, Finance Committee, National Academy of Sciences
4. Member, COSEPUP (Committee of Science, Engineering, and Public Policy) of the National Academy of Sciences

5. Member, Physics Policy Committee, American Physical Society
6. Principal Editor, Journal of Materials Research
7. Editor (Physics, Materials Science), Science Spectra
8. Member, Advisory Committee of Engineering School, University of California, Berkeley
9. Member, Miller Institute Advisory Board, University of California, Berkeley
10. Member, Selection Committee for START and Wittgenstein Awards, Austrian Science Foundation (Vienna)
11. Member, Packard Award Selection Committee

## **NEW DISCOVERIES, INVENTIONS OR PATENT DISCLOSURES**

None.

## **HONORS AND AWARDS**

1. Chairman of the Board, American Association for the Advancement of Science
2. Doctor of Science Honoris Causa, Ohio State University, June 1998
3. Hunter College Award for Outstanding Professional Achievement, 1998
4. Hall of Fame Award, WITI (Women in Technology International), 1998